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Using laser pulses of duration comparable to semiconductor relaxation and dephasing times, the coherent phenomena common to two-level systems have been demonstrated in II-VI and III-V semiconductors. These light-matter interactions are characterized by electrons and holes in their initially excited states and/or a macroscopic polarization that persists after the excitation.

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STATEMENT OF PROBLEM STUDIED

The steady-state optical absorption of matter comes from a balance between the dynamic processes of optical excitation and several decay mechanisms. When pump and probe experiments are conducted with laser pulses shorter than these decay processes, a vast body of coherent effects is opened up. Experimentally, coherent effects are difficult to observe in semiconductors because of very fast dephasing times, on the order of picoseconds for excitons,[1] and tens of femtoseconds for band-to-band transitions.[2]

These effects have been attacked in the last decade with femtosecond lasers and more sophisticated theoretical models. This project was directed toward nonlinear absorption dynamics in the sub-T_1 and sub-T_2 time regions. Part 1 used spectral hole burning to investigate nonthermal carrier distributions in bulk and quantum dot CdSe. The questions that were to be answered were: Since intraband scattering is the fastest source of population and polarization decay in bulk semiconductors, how would this change in 3D quantum confined semiconductors? That is, what states are observed in quantum dots, and can quasi-steady-state hole-burning take place?

Parts 2 and 3 both used sub-T_2 pulses and ultrafast nonlinear effects in semiconductors to investigate a macroscopic polarization that persists longer than the optical pulses. Part 2 addressed the optical Stark effect in excitons. Specifically, three points were to be addressed:

1) How closely does the shift follow the pump intensity? Much of the initial interest in the exciton shift came from the need for optical switching devices whose switch-off time is not carrier lifetime dependent. The OSE appeared to provide for a shift only as long as the field is present. The first experiments[3-5] showed femtosecond or picosecond recovery times, but the time resolution was insufficient to give a clear

answer. In particular, it was unknown how these results change when the time resolution becomes shorter than the dephasing time.

2) How much does the exciton shift? Closely related to the question of shift dynamics, is that of the magnitude of the shift. The previous observations mentioned above were inconclusive about the agreement between measured and calculated shift. A sub-T_2 pump and probe experiment violates the intuitive interpretation of measuring the instantaneous effects of the pump. Thus, there was expected to be disagreement between the measured shift and the results of a steady-state calculation that employs the peak (fs) field as a cw field.

3) Can a pure shift be obtained? Two distinct effects have tended to obscure observation of the OSE. Real carrier generation is always present to some degree, either from off-resonant linear absorption or from two-photon absorption. This produces bleaching and broadening of the exciton after the arrival of the pump pulse. These effects are well understood, but undesirable. In addition to hindering observation, this is troublesome from the standpoint of designing ultrafast recovery switches. Thus there was interest in finding experimental conditions that minimize these effects.

The second effect that departs from a pure shift is *transient* modification of the exciton oscillator strength. Although the nonresonant absorption is described in terms of virtual carriers, they can be thought of as very real carriers that exist during the short time of the pump pulse. During this transient time, they can have all the many-body effects on the exciton absorption that real carriers have.

In part 3, spectral oscillations in the differential absorption spectra were discovered in several sub-T_2 pump and probe experiments. These were further investigated to see under what conditions they occur, and their precise physical origin.

SUMMARY OF IMPORTANT RESULTS

The goals to be studied required a particularly specialized laser system that was not yet commercially available: tunable $\simeq$ 100 fs pump pulses and broadband fs probe pulses. This was achieved by constructing a colliding-pulse modelocked oscillator, amplifying the output to μJ energies, generating a white light continuum, and then reamplifying spectral portions of the continuum. The finished system is capable of exciting semiconductor excitons and band states throughout the yellow to NIR. This design is covered in Appendix A.

Using laser pulses of duration comparable to semiconductor relaxation and dephasing times, the coherent phenomena common to two-level systems have been demonstrated in II-VI and III-V semiconductors. These light-matter interactions are characterized by electrons and holes in their initially excited states and/or a macroscopic polarization that persists after the excitation.

A nonthermal carrier distribution was observed in bulk CdSe, demonstrating the extremely fast energy relaxation possible when carrier-LO-phonon scattering is included.[6] Quantum-confined CdSe microcrystallites were then employed to limit the available decay avenues. The one and two-pair transitions were characterized,[7] and using spectral hole-burning, energy relaxation was shown to be substantially slower than in bulk. Dipole dephasing remained very rapid.

Persistent macroscopic polarization was demonstrated in semiconductors through pump and probe experiments conducted on time scales faster than the inverse linewidth of the exciton. In the first set of experiments, it was found that the exciton optical Stark shift deviates from that predicted by a steady-state theory. The shift is smaller than expected and lasts longer than the pump pulse.[8-9] Both effects are explained by free evolution of the exciton's polarization. Transient exciton bleaching is observed and shown to be a result of adiabatic following.

In a second study of coherent polarization, spectral oscillations at negative delay times were studied. Oscillations were measured in a wide variety of semiconductors, temporally preceeding several different pump-probe phenomena.[10-11] By comparison with a semiclassical theory, it was shown that spectral oscillations in the spectral region of the exciton are evidence of persistent exciton polarization. Oscillations in the band are due to four wave mixing of the pump and probe beams.

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LIST OF PERSONNEL

Brian Fluegel: Ph.D. granted 5/14/92.

Dissertation: "Sub-relaxation and sub-dephasing dynamics of light-induced polarization in semiconductors."

Dissertation advisor: Nasser Peyghambarian

REPORT OF INVENTIONS

No inventions reported

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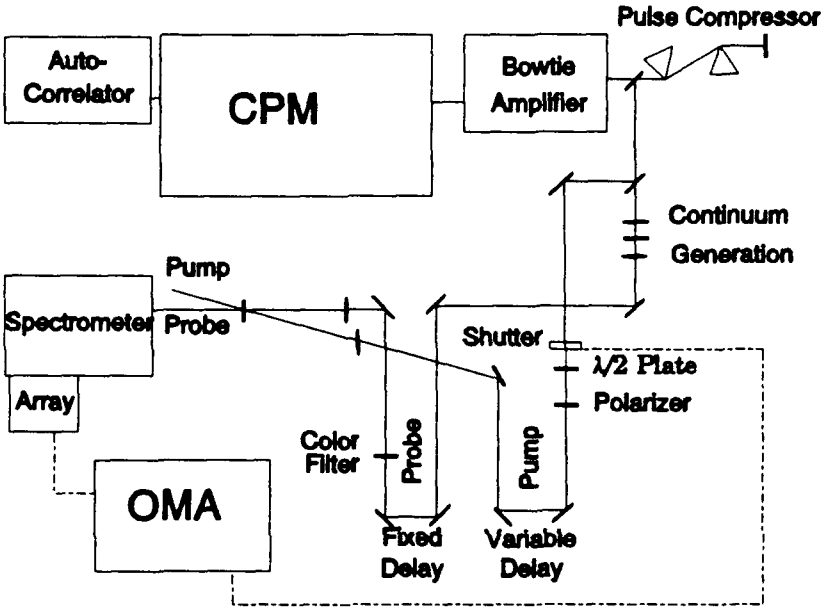
APPENDIX A - LASER SYSTEM

The general method to be used for observing sub-T_1/T_2 effects in semiconductors is femtosecond transient absorption measurements. Fig. A-1 outlines the major



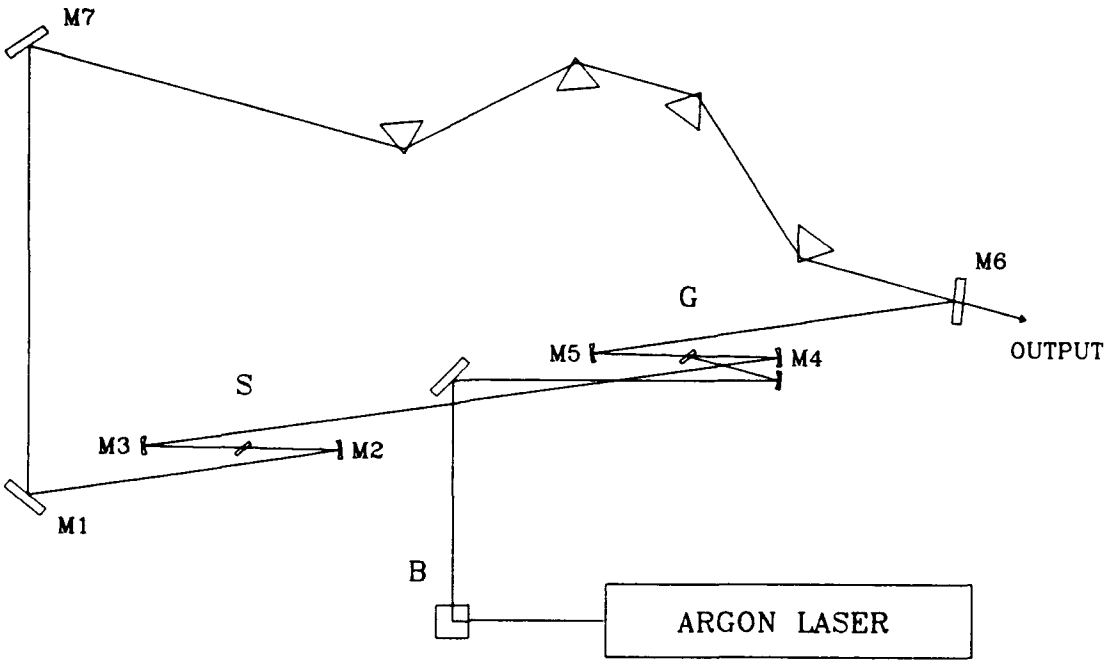
A-1 Schematic diagram of transient absorption measurement.

components: an ultrafast laser system, a pump-probe setup, and a spectrometer/OMA. Since the measurements' temporal resolution is determined by the laser pulsewidth, the design of the laser receives special attention.

Oscillator

The resonator used here is the colliding-pulse modelocked (CPM) ring dye laser, and differs very little from the original designs of Fork et al.[12] and Valdamis et al..[13] The salient features of this oscillator (Fig. A-2) are as follows.

A cw argon laser pumps a jet of rhodamine 6G. It is the (relatively) long storage time of this dye which allows cw pumping to provide enough inversion. The convenience and simplicity of this type of laser over synch-pumps cannot be



A-2 Colliding-pulse modelocked ring-dye laser. M1 and M2 have 25 mm radius of curvature, M4 and M5 have 50 mm radius. S = saturable absorber jet, G = gain jet. M6 is the 3% output coupler, B rotates the polarization by beamsteering. Prisms are fused silica with a slant length of 29 cm.

overstated. Passive modelocking occurs in a slow saturable absorber (SA), a thin jet of DODCI, labeled S in Fig. A-2. With no wavelength selecting elements, lasing occurs at the overlap of R6G gain and the DODCI transmission, about 620 nm.

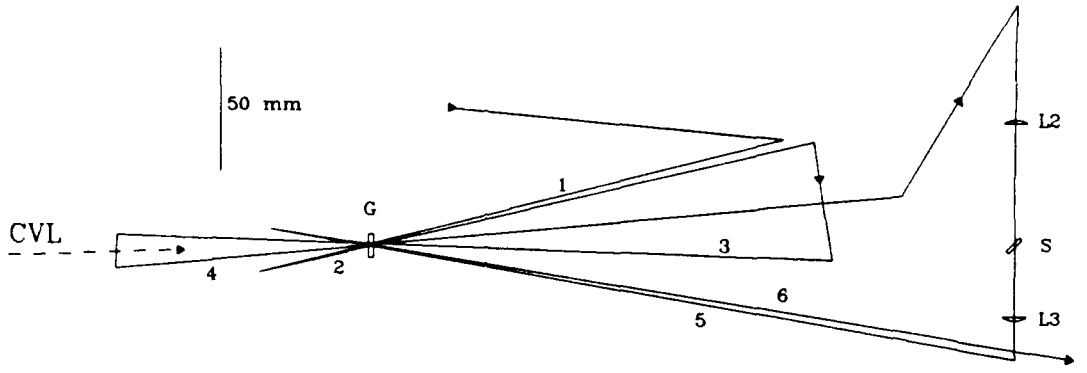
The stability of this modelocking is enhanced by the bi-directional ring cavity. Minimum round-trip loss occurs when two counterpropagating pulses collide at the SA. The two pulses form a grating which i) increases the saturation, and ii) scatters light from one direction into another, thus keeping the two beams coupled and coherent.[14] Forming a counterpropagating grating with fs pulses requires a thin (35 μm) jet.

Sub-picosecond pulses are possible because of efficient saturable gain and absorption, a large bandwidth, and a minimum of dispersive optics. With the addition of Brewster prism compensation of group velocity dispersion[15] it is possible to

routinely achieve 60 fs pulses, with 10 mW of output power.

Dye Amplifier

As seen in Fig. A-1, the output of the CPM is sent directly into a copper vapor laser (CVL)-pumped dye amplifier. This is a much modified version of the original



A-3 Scale diagram of 620 nm dye amplifier. S = saturable absorber jet, G = gain cell, N (1...6) = approach to Nth pass. L2 and L3 are focussing and collimating lenses for the satuarble absorber. The input CPM beam is coupled by L1 = 1000 mm (not shown), and the CVL is focussed by L4 = 600 mm.

six-pass "bowtie" design.[16] Fig. A-3 shows a scale drawing of the optical path. All six passes are in one plane, and conventional mirror mounts are used, but the mirrors have been positioned as close as possible to minimize the angles between passes. This makes the amplifier more longitudinally pumped and improves the spatial beam quality. The usual refocussing lens between passes 2 and 3 has been eliminated. This enables the mirrors to be positioned even closer, removing the need for refocussing, since lens 1 alone can produce 1 to 1.5-mm diameter spots in the first four passes.

A single 300-μm thick saturable absorber of Malachite Green in ethylene glycol between the fourth and fifth passes reduces amplified spontaneous emission (ASE). Two mirrors have been used for the input to the SA. This is an additional loss, but permits the pass angles to be kept still smaller. The gain cell flows cooled Rhodamine

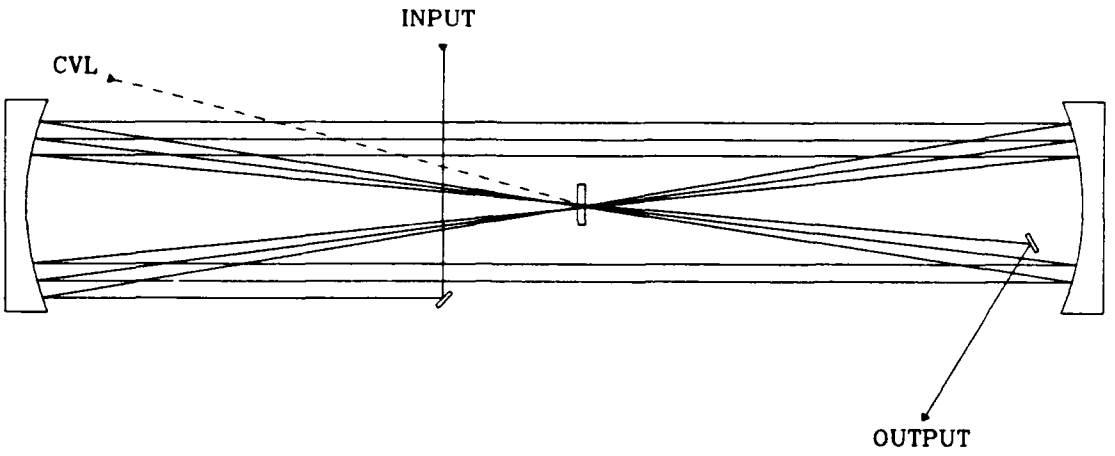
640 in ethylene glycol. This cell is 2 mm thick, fused silica, with in-line hose nipples, and anti-reflection coated for 620 nm. Using a dye cell for the gain stage gives output a factor of 2 higher over that from a dye nozzle, along with increased stability. The increased group velocity dispersion can still be compensated by the prism compressor, yielding 60 fs pulses. This prism pair consists of SF10 equilateral prisms with a spacing of 30 cm. At this point, the pulse energy is 2-10 μJ depending on CVL power.

The CVL benefits from the Metalaser Technologies short pulse apparatus. This consists of a polarizing beamsplitter cube in the cavity which reflects the vertical polarization out of the cavity, through an eight-foot delay, and back into the cavity. This very large feedback is timed to deplete the gain, thus truncating lasing in the horizontal polarization after a single round-trip oscillation. The output is 10 W average power, horizontally polarized, in a 10-nsec pulse. The beam is focussed to a 2-mm diameter spot using a 600-mm lens. Operation with the short pulse apparatus causes the amplifier output to be reduced by $\simeq$ 20%, but ASE is reduced by a factor of 10 with no amplification of a second CPM pulse.

Several of the planned experiments will use a 620 nm pump. In these cases, a portion of the laser is split off just after the prisms. It then passes through a shutter, a precision optical delay, and is rotated to vertical polarization. The remainder of the 620 nm beam is focussed onto a 2-mm thick jet of ethylene glycol to generate a broadband continuum probe.[17]

Continuum Amplification

For some experiments, a tunable pump is required; continuum reamplification (not shown in Fig. A-1) is used here. A part of the continuum is split off and filtered with a 10-nm bandpass interference filter centered around the desired pump wavelength. The narrowband output is then coupled into the confocal amplifier shown in Fig. A-4.



A-4 Confocal amplifier for continuum reamplification. Concave mirrors have radii of curvature 240 mm and 300 mm. By translating the exit mirror, the number of passes may be easily varied.

This design is simpler to build and operate than the amplifier of Fig. A-3, however there is no provision for a SA. Thus the bowtie design is still prefered for CPM amplification because its SA allows higher pumping which produces larger pulse energies. For continuum reamplification however, the performance requirements are lower and suitable saturable absorbers are not available at all wavelengths. For these reasons the confocal amplifier of Khoroshilov et al.[18] was chosen.

As seen in Fig. A-4, this consists of two unequal radii concave mirrors whose focal points coincide at the gain jet. The collimated continuum beam enters from one side, and is repeatedly focussed onto the jet, then recollimated closer to the axis. The gain jet is a 1.3-mm thick sapphire nozzle using a solvent of ethylene glycol and glycerol. This is pumped by a CVL of $\simeq$ 15 W average power focussed to a 1 to 2-mm diameter spot. Depending on the dye used, either the yellow (578nm) or green (510 nm) line may be filtered out before reaching the gain jet. After six passes, a small exit mirror picks the beam out of the cavity. The pulses are then recompressed in a prism pair similar to those in Fig. A-1. Pulsewidths as short as 130 fs and energies up to 500 nJ

are obtained, depending on wavelength. By changing the dye and interference filter, the amplifier has been used from 550 nm to 830 nm

The actual experiment is a time resolved pump and probe measurement. The pump and probe are focussed to $\simeq$50 μm onto a sample held in a closed-cycle liquid helium cryostat. The transmitted probe is sent to a spectrometer and OMA. Pump and probe are orthogonally polarized, and nearly colinear, separated by about 15°. By convention, a time axis is defined with the pump pulse arriving at the sample at time t=0. The probe arrives at time t=t_p. Thus for positive t_p, the pump arrives first.

Data Acquisition

For effective data acquisition, the laser noise spectrum should be analyzed, and an appropriate modulation frequency chosen. In this laser, it is found that noise is essentially flat above a few 10's of Hz, and rises sharply below. Rising noise at low frequencies is typical of many physical systems and for this reason it is best not to make simple "DC" measurements. For this laser system, the modulation should avoid the $\leq$ 10 Hz region.

This is difficult in an optical absorption measurement, since it would require positioning and repositioning the sample at 10 Hz, sometimes with 25 μm accuracy. The technique used instead is to rely on differential absorption, a measurement which does not require moving the sample. Here we measure: absorption in the presence of pump minus absorption without pump.

This is accomplished by programming the OMA to compute:

$$ln\left(\frac{I_{probe}(\text{Pump on})}{I_{probe}(\text{Pump off})}\right) = ln\left(\frac{I_{in}e^{-\alpha L}}{I_{in}e^{-\alpha_0 L}}\right) = ln(e^{-(\alpha-\alpha_0)L}) = -\Delta\alpha L \qquad (A.1)$$

where I_{probe} is the probe beam transmitted through the sample, L is the sample thickness, and a subscript 0 means the linear, unexcited state. The OMA switches the pump on and off at 10 Hz using an electrical shutter, while I_{probe} is synchronously

accumulated in two separate data bins: one for pump-on, one for pump-off. Thus the OMA simulates a lock-in amplifier with 1024 channels. In some experiments the results are calculated in the related form of differential transmission spectra (DTS) defined by:

$$\text{DTS} \equiv \frac{T-T_0}{T_0} = \frac{I_{in}e^{-\alpha L} - I_{in}e^{-\alpha_0 L}}{I_{in}e^{-\alpha_0 L}} = e^{-(\alpha-\alpha_0)L} - 1 \qquad A.2$$

For small signals this is equivalent to $-\Delta\alpha L$. Regions of positive signal correspond to bleaching while negative signal indicates induced absorption.

In certain situations the spectral data must be corrected for chirp: group velocity dispersion of the probe beam. This arises from the ethylene glycol jet and subsequent optics. Because of this, a DTS spectrum at nominal delay time τ will consist of data from $t>\tau$ at the blue wavelengths, and data from $t<\tau$ at red wavelengths. If one only concentrates on a narrow band of wavelengths during the experiment, the chirp can be ignored. For broadband data, the chirp must be corrected. This is done by first measuring the chirp using sum frequency generation and then taking experimental data at very short time delays. A computer then takes the set of chirped curves, and using the measured value of the chirp, interleaves the data into a new set of dechirped files. Linear interpolation is used for time delays between the original files. Since the original files consist of data skewed in time, the processed files will have blank regions at the extreme time delays curves.